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A Compilation of Thermodynamic and Transport Properties of Aqueous Sulfuric Acid

Bert R. Staples and Theresa F. Wobbeking

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U.S. DEPARTMENT OF COMMERCE, Malcolm Baldrige, *Secretary*
NATIONAL BUREAU OF STANDARDS, Ernest Ambler, *Director*

A COMPILATION OF THERMODYNAMIC AND
TRANSPORT PROPERTIES OF
AQUEOUS SULFURIC ACID

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ABSTRACT

A detailed compilation of sources of data for the thermodynamic and transport properties of aqueous sulfuric acid is presented. All ranges of temperature, concentration, and pressure are included.

Key Words: Activity coefficients; aqueous; compilation; conductivity; enthalpy; electrolytes; Gibbs energy; osmotic coefficients; solutions; sulfuric acid; thermodynamic properties; transport properties.

Appendix 1. Annotated Portion

Abel, E., J. Phys. Chem., 50, 260 (1946).

Represents vapor pressures of H_2SO_4 from 2.5 to over 90 mol/kg.

Bass, S. J., Gillespie, R. J., Oubridge, J. V., J. Chem. Soc., 837, (1960).

The authors have measured the osmotic coefficients of aqueous H_2SO_4 for compositions of 0.00 to 0.40m.

Burt, B. C., J. Chem. Soc., 85, 1339 (1904).

Reports the vapor pressure of sulfuric acid solutions for temperatures from 55 to 235C and for concentrations ranging from 24.92 to 95.94% H_2SO_4 .

Collins, E. M., J. Phys. Chem., 37, 1191 (1933).

The vapor pressures for sulfuric acid solutions was obtained for temperatures from 24.87 to 135.61C and for concentrations ranging from 29.90 to 65.47% H_2SO_4 .

Covington, A. K., Dobson, J. V., and Wynne-Jones, Trans. Far. Soc., 61, 2050 (1965).

Report data for the composition range of 0.007 to 0.1 mol/kg and 5 data sets for the cell Pt; H_2 , H_2SO_4 (m), $PbSO_4$, PbO ; Pt, 13 data sets for the cell Pt; H_2 , H_2SO_4 (m), Hg_2SO_4 , Hg ; Pt and 7 points for the cell Pt; H_2 , H_2SO_4 (m), Ag_2SO_4 ; Ag.

Daudt, W., Z. Physik. Chem., 160, 255 (1923).

Measured vapor pressures at very low temperatures for compositions of 20-60 mol/kg H_2SO_4 .

Deno, N. C. and Taft, R. W., Jr., J. Am. Chem. Soc., 76, 244 (1954).

Used the Hammett relation to calculate the activity of water in aqueous H_2SO_4 at molalities exceeding 30 mol/kg.

Duisman, J. A. and Giauque, W. F., J. Phys. Chem., 72, 562 (1968).

Used a variety of data sources to derive thermodynamic quantities for the lead storage cell and a set of emf values from third law considerations.

Edward, J. T. and Wang, I. C., Can. J. Chem. 43, 2867 (1965).

Measured the dissociation of the bisulphate ion over a concentration range of 0.010 to 0.30 OM at an temperature of 27.0+/-1.5C.

Feilchenfeld, H. and Fuchs, J., Israel J. Chem., 12, 899 (1974).

Measured the hydration of H_2SO_4 at 25C.

Flis, I. E., Mischenko, K. P., Pakhomova, N. Y., Russ. J. Inorg. Chem., 3, 1772 (1958).

The authors have measured the heat of neutralization of H_2SO_4 at 10, 25, 35 and 50C. The dissociation of the bisulphate ion was calculated at temperatures of 10, 25, 35, and 50C.

Gardner, W. L., Jekel, E. C. and Cobble, J. W., J. Phys. Chem. 73 (6), 2017 (1969). The standard partial molal heat capacities of H_2SO_4 have been obtained by the integral heat method between 0 and 100C.

Gardner, W. L., Mitchell, R. E., and Cobble, J. W., J. Phys. Chem. 73 (6), 2021 (1969).

The authors used the third law method to calculate a set of activity coefficients as a function of temperature (0 to 55C) and concentration (0.1 to 4.0 mol/kg).

Glueckauf, E., and Kitt, G. P., Trans. Far. Soc. 52, 1074 (1956).

Used a bithermal isopiestic method to determine osmotic coefficients between 20 and 65 mol/kg. These authors added a constant correction of 0.08 in the values of ($m_2 - m_1$) to bring their results in agreement with others. The Clausius-Clapeyron relationship was used to construct a curve of osmotic coefficients at 25C.

Groenier, W. L., Partial dissertation, The University of Chicago (1933).

The heats of dilution of H_2SO_4 solutions at approximately 25C have been measured over a concentration range of about 0.003 to almost 4.0m.

Grollman, A. and Frazer, J. C. W., J. Am. Chem. Soc., 47, 712 (1925).

Determined the vapor pressures of aqueous sulfuric acid solutions at 25C. Thirteen values of the osmotic coefficient have been calculated from these data. The molalities range from 0.1 to 3 mol/kg.

Hamer, W. J., J. Am. Chem. Soc., 57, 9 (1935).

Determined the emf of the cell Pt; H_2 , H_2SO_4 (m), $PbSO_4$; Pt for compositions of 0.005 to 7 mol/kg over a temperature range of 0-60C.

Hamer, W. J. Am. Chem. Soc., 57, 662 (1935).

Made measurements of the cell Hg ; Hg_2SO_4 , H_2SO_4 (m), Hg_2SO_4 , Hg over an concentration range of 0.05 to 17.0 molal and at the temperatures of 0, 10, 15, 25, 35, 45, and 60C. The transference numbers of the hydrogen ion of sulfuric acid were calculated over an concentration range of 0.05 to 17 molal and at temperatures of 0, 10, 15, 25, 35, 45, and 60C.

Harned, H. S. and Hamer, W. J., J. Am. Chem. Soc., 57, (1935).

Reported the emfs of the cell Pt; H_2 , H_2SO_4 (m), Hg_2SO_4 , Hg ; Pt over a concentration range of 0.05 to 17.5 mol/kg at temperatures from 0 to 60C.

Hausrath, H., Ann. Phys. (Leipzig) 9, 522 (1902).

The freezing point method was used for 8 molalities in the dilute range from 0.0001 to 0.02 mol/kg.

Hepburn, J. R. I., Proc. Phys. Soc. London, 40, 249 (1928).

Determined the vapor pressures of H_2SO_4 solutions using a dew point method for 9 solutions from 7 to 12.5 mol/kg.

Hornung, E. W., and Giauque, W. F., J. Am. Chem. Soc., 77, 2744 (1955).

Performed a direct set of vapor pressure measurements on 3 solutions (14-28 mol/kg) as a function of temperature.

Jones, F. R., J. Appl. Chem. 1, S144 (1951).

An absorption method was employed by Jones to calculate the vapor pressure of H_2SO_4 at rounded molalities from 0.5 to 55 mol/kg.

Jones, H. C. and Getman, F. H., Am. Chem. J., 27, 433 (1902).

Have reported freezing point measurements for 9 concentrations (0.1 to 2.8 mol/kg).

Jones, H. C., Getman, F. H., Bassett, H. P. McMaster, L. and Uhler, H. S., Carnegie Institution of Washington, Publication No. 60, Washington, D.C. 1907.

Have reported freezing point measurements for 16 molalities between 0.1 and 6.4 mol/kg.

Jones, H. C., Z. Phys. Chem. (Leipzig) 12, 623 (1893).

Has reported freezing point measurements for 11 dilution solutions between 0.001 to 0.1 mol/kg.

Kerker, M., J. Am. Chem. Soc., 79, 3664 (1957).

Measured the quotient of activity coefficients of H_2SO_4 for compositions ranging from 0 to 1.80m at temperatures of 18, 25, and 50C. The dissociation of H_2SO_4 was measured at 18, 25, and 50C for concentrations of 0.0005 to 1.80m. The equivalent conductance of HSO_4^- was measured from 0.0 to 0.5m at temperatures of 18, 25, and 50C.

Kunzler, J. E. and Giauque, W. F., J. Am. Chem. Soc., 74, 5271 (1952).

Freezing point depressions were measured but all molalities were above 30 mol/kg.

Lange, E., Monheim, J. and Robinson, A. L., J. Am. Chem. Soc., 55, 4733 (1933).

Measured the heat of dilution of aqueous H_2SO_4 for concentrations of 0.00005 to 0.5m at an temperature of 25C.

Lilley, T. H. and Briggs, C. C., Electrochim Acta, 20, 257 (1975).

Empolyed earlier emf data (Shrawder and Cowperthwaite, 1934) to calculate activity coefficients in the dilute region of 0.001 to 0.02 mol/kg.

Loomis, E. H., Ber Bunsenges. Phys. Chem., 26, 797 (1893).

Five freezing points were reported. The molalities for these points range between 0.01 to 0.2 mol/kg.

MacDougall, F. H. and Blumer, D. R., J. Am. Chem. Soc., 55, 2236 (1933).

The emf of the cell Pt; H_2 , H_2SO_4 (m), $HgSO_4$, Hg; Pt was measured with varying additions of acetic acid. The authors also report measurements for 7 solutions where no acetic acid was added. These molalities varied from 0.05 to 2.2 mol/kg.

McHaffie, I. R., J. Chem. Soc., 112 (1927).

Determined vapor pressures for high molalities (above 20 mol/kg) using a vapor condensation method.

Morgan, J. J., Bender, D. A. and Capell, R. G., Chem. Met. Eng., 50, 122 (1943).

The authors measured the specific heats of H_2SO_4 at 80C.

Pickering, S. U., Ber. Dtsch. Chem. Ges., 25, 1099 (1892).

About 40 measurements of freezing points of H_2SO_4 solutions ranging from 0.001 to 0.4 mol/kg were made.

Pitzer, K. S., J. Phys. Chem., 80, 2863 (1976).

Has taken into account the dissociation of the HSO_4^- ion to describe some of the thermodynamic properties of sulfuric acid solutions.

Pitzer, K. S., Roy, R. N., and Silvester, L. F., J. Am. Chem. Soc., 99, 4930 (1977).

Calculated the cell potentials for H_2SO_4 at approximately 25C for compositions of 0.005 to 1.0 m. The activity and osmotic coefficients were determined for compositions of 0.1 to 6.0 m at approximately 25C.

Quist, A. S., Marshall, W. L., Jolley, H. R., J. Phys. Chem., 69, 2726 (1965).

The electrical conductances of dilute sulfuric acid solutions were measured at temperatures from 0 to 800 C and at pressures from 1 to 4000 bars. The first ionization constant of H_2SO_4 was calculated at densities below $0.8g/cm^3$ and at temperatures between 400² and 800 C. The second ionization constant of H_2SO_4 was calculated at temperatures from 100 to 300 C and at densities up to $1.0g/cm^3$.

Randall, M. and Cushman, O. E., J. Am. Chem. Soc., 40, 393 (1918).

Utilizing the cell $Pt; H_2, H_2SO_4 (m), Hg_2SO_4, Hg; Pt$, Randall and Cushman measured the free energy of dilution for 7 mole fractions corresponding to 0.005 to 8.2 mol/kg. The results were given only to the nearest 0.1mV.

Randall, M. and Scott, G. N., J. Am. Chem. Soc., 49, 647 (1927).

Have determine freezing points of 33 solutions from 0.001 to 0.1 mol/kg.

Randall, M. and Taylor, M. D., J. Phys. Chem., 45, 959 (1941).

Measured the heat capacity of aqueous H_2SO_4 for compositions of 0.05 to 2.4 m.

Rard, J. A. and Miller, D. G., Unpublished Data, (1977).

Have made available unpublished isopiestic data using KCl as a reference. The molalities range from 0.1 to 3 mol/kg. for 17 points.

Rard, J. A. and Spedding, F. H., J. Chem. Eng. Data, 22, 56 (1977).

Have carefully determined the isopiestic molalities of 60 solutions of H_2SO_4 using $CaCl_2$ is a reference. Their equilibration times of 4 days or more were generally double those by previous investigators. The molalities range from 3.8 to 13.3 mol/kg. and the measurements were taken at 25C.

Robinson, R. A., Trans. Faraday Soc., 35, 1229 (1939).

The activity coefficient of sulfuric acid has been determined by the isopiestic method at 25 C between 0.2 and 3 mol/kg. Robinson has used KCl as a reference to determine the isopiestic ratios of 53 solutions from 0.2 to 3 mol/kg.

Robinson, R. A., Trans R. Soc. N. Z., 75, 203 (1945).

Determined the isopiestic ratios of $H_2SO_4/NaCl$ at 25C.

Roth, W. A., and Knothe, W., Unpublished data given in Landolt-Bornstein Physikalisch-Chemische Tabellen, Auflage 6, Band II Teil 2, p. 862, A. Eucken, Editor, Springer-Verlag, Berlin (1960). Have determined freezing point depressions at 3 molalities below 0.1 mol/kg.

Scatchard, G., Hamer, W. J., and Wood, S. E., J. Am. Chem. Soc., 60, 3061 (1938). Determined the isopiestic ratios of H_2SO_4 and both NaCl (32 points) and KCl (23 points), from 0.1 to 4.5 mol/kg.

Shankman, S. and Gordon, A. R., J. Am. Chem. Soc., 61, 2370 (1939). The vapor pressure of sulfuric acid solutions at 25°C has been measured by the static method for concentrations ranging from 2 to 23 molal.

Shrawder, J. Jr., and Cowperthwaite, I. A., J. Am. Chem. Soc., 56, 2340 (1934). Have determine the activity coefficients of H_2SO_4 as a function of temperature for 5 dilute solutions (0.001 to 0.02 mol/kg). They employed the following cell Pb-Hg (2 phase), $PbSO_4$, H_2SO_4 (m), H_2 , with an lead amalgam electrode.

Stokes, R. H., J. Am. Chem. Soc., 67, 1686 (1945). The activities and partial molal heat contents of water in sulfuric acid solutions at 25°C were computed over a concentration range of 0.01 to 17m directly from the electromotive forces of cells without transference.

Stokes, R. H. J. Am. Chem. Soc., 67, 1689 (1945). Sodium hydroxide was used as a reference in the isopiestic determinations for 33 data sets of H_2SO_4 from 1.6 to 21.6 mol/kg at 25°C.

Stokes, R. H., Trans. Far. Soc., 41, 637 (1945). Measured the isopiestic molalities of H_2SO_4 and $CaCl_2$ at 25°C. The 32 molalities of H_2SO_4 ranged from 4.3 to 15.4 mol/kg.

Stokes, R. H., Trans. Far. Soc., 44, 295 (1948). The osmotic and activity coefficients of H_2SO_4 were measured at a temperature of 25°C over a concentration range of 0.1 to 5.0m.

Trimble, H. M. and Ebert, P. F., J. Am. Chem. Soc., 55, 958 (1933). Report emf measurements of the cell Pt; H_2 , H_2SO_4 (m), Hg_2SO_4 , Hg; Pt with ethylene glycol additions. The authors also reported measurements for 6 compositions from 0.005 to 1 mol/kg, for which there was no ethylene glycol added.

Turner, David J., J. Chem. Soc. Faraday Trans. I, 70, 1346 (1974). The dissociation of the bisulphate ion was measured at 25°C for a concentration range of 0.020 to 4.00m.

Vdovenko, V. M. Lazarev., L. N. and Khvorostin, Ya. S., Russian J. Inorganic Chem., 12, 610 (1967).

Dissociation constants of HSO_4^- at ionic strengths of 0.50, 1.00, and 2.00 have been determined at temperatures of 20.0, 25.0 and 35°C.

Vosburgh, W. C. and Craig, N., J. Am. Chem. Soc., 51, 2009 (1929). Calculated and measured the potentials of the cell Pt; $PbSO_4$, PbO_2 , H_2SO_4 (m) / H_2SO_4 (m), Hg_2SO_4 ; Hg. The emfs of solutions from 0.05 to 3.5 mol/kg were measured at 25°C.

Wilson, R. E., Ind. Eng. Chem., 13, 326, (1921).

Measured the vapor pressure of sulfuric acid solutions for temperatures from 0 to 75C and for concentrations ranging from 0.0 to 80% H_2SO_4 . Wilson also reports the heats of dilution of H_2SO_4 for temperatures from 12.5 to 62.5% H_2SO_4 .

Wirth, H. E., Electrochim. Acta, 16, 1345 (1971).

Presents a summary of emf measurements of H_2SO_4 solutions and uses the dissociation of the bisulfate ion to calculate activity coefficients.

Wu, Y. C. and Young, T. F., Enthalpies of Dilution of Aqueous Electrolytes:

Sulfuric Acid, Hydrochloric Acid, and Lithium Chloride (Paper is in press). Wu and Young conducted measurements of the enthalpies of dilution of aqueous H_2SO_4 at 25C for compositions of 0.00090 to 6.4 mol/kg.

Appendix 2. Bibliography

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"The Behavior of the Lead Dioxide Electrode."
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"The Depression of Freezing Point in Very Dilute Aqueous Solutions."
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